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# Simultaneous Estimation of Particle Size, Relative Refractive Index and Concentration of Latices by Turbidity Measurements (Special Issue on Polymer Chemistry X)

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## Simultaneous Estimation of Particle Size, Relative Refractive Index and Concentration of Latices by Turbidity Measurements

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A method to estimate simultaneously the size and concentration of particles in latices and the relative refractive index of those to the surrounding medium by turbidity measurements at three different wavelengths is proposed. The values obtained for polystyrene latex by this method agreed well with those obtained by other methods. The validity of this method is discussed.

### INTRODUCTION

Light transmission as well as light scattering offer various methods for characterizing colloiddally dispersed particles. The theory for such methods is given by applying the Mie equation to the light scattering for an isotropic spherical particle.

According to the theory, the turbidity  $\tau$  of latices is expressed as,

$$\tau = NSK$$

Here the particles in the latices are assumed to be sphere of equal size.  $K$  is a scattering coefficient derived from the Mie equation,  $N$  is the number of spheres in unit volume (the concentration of latices) and  $S$  is the geometrical cross-section of the sphere ( $\pi r^2$ ,  $r$ : the radius of sphere).  $K$  is a function of the radius ( $r$ ), the refractive index of sphere relative to surrounding medium ( $m$ ) and the wavelength of incident beam in surrounding medium ( $\lambda^*$ ). Therefore,  $\tau$  is a function of  $N$ ,  $r$ ,  $m$  and  $\lambda^*$ .

Consequently, when  $N$  and  $m$  are known,  $r$  can be evaluated from  $\tau$  at a given  $\lambda^*$ , with the numerical computation of the scattering coefficient  $K$ . This method has been applied to a system containing monodisperse particles, such as polystyrene latex, by Tabibian, Heller and Epel,<sup>1)</sup> Dezelic and Kratochvil and Maron, Pierce and Ulevith.<sup>3)</sup>

Secondly, when only  $m$  is known,  $r$  and  $N$  can be evaluated from the turbidities at two different wavelengths with a similar computation of  $K$ . According to the foregoing equation, the ratio between the turbidities at two given  $\lambda^*$  is equal to the ratio between values of  $K$  at each  $\lambda^*$ . Thus, the ratio of  $\tau$  at two given  $\lambda^*$  is a function of  $r$  alone for a given  $m$ . This ratio has been called the "dispersion quotient" (denoted as DQ) by Teorell.<sup>4)</sup> He suggested that the DQ was to be related to particle size of colloidal dispersion. Evva<sup>5)</sup> applied this DQ-method to colloidal dispersion of silver halides. Sakurada, Hosono and Tamamura<sup>6)</sup> has verified the validity of this relation by comparing

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the radii of various synthetic latices calculated from the DQ values with those obtained by electron microscopy. Another check of this method has been carried out for polystyrene latices by Deželić, Deželić and Tažák.<sup>7)</sup>

Finally, if all of  $r$ ,  $c$  and  $m$  are unknown, these quantities should be able to be evaluated from turbidities at three or more different  $\lambda^*$ , because as mentioned above  $\tau$  is a function of the four variables; that is,  $r$ ,  $c$ ,  $m$  and  $\lambda^*$ .

In the present paper, it is intended to propose a method to estimate the particle size, the concentration and the relative refractive index of latices from turbidity ratios at three or more different wavelengths and discuss the validity of this method.

## THEORETICAL

### (a) Relationship between Turbidity and Scattering Coefficient

A light beam traversing a scattering system containing  $N$  spheres per unit volume is being attenuated, the attenuation being equal to the total scattered radiation; *i.e.*,

$$\frac{dI}{I dx} = NR \quad (1)$$

and, for a finite optical path,  $x=l$

$$\ln I_0/I = \ln(T)^{-1} = NRl \quad (2)$$

where  $I_0$  is the intensity of the incident beam,  $I$  the intensity of the beam emerging from the scattering solution at  $x$ ,  $R$  the coefficient of apparent absorption resulting from light scattering of one sphere and  $T$  the light transmission.

Generally  $NR$  is designated by a symbol

$$\tau = NR \quad (3)$$

denoted as "turbidity". The scattering coefficient  $K$  is defined as the ratio of the total outward flux of energy in the scattered wave to the flux of energy incident on the sphere in the incident wave. In other words,  $K$  is defined as the ratio of the scattering cross-section to the geometrical cross-section of the sphere. Accordingly,  $K$  is represented as

$$K = R/S \quad (4)$$

Here  $S$  is the geometric sectional area ( $\pi r^2$ ,  $r$ : the radius of the sphere). Eventually, the turbidity  $\tau$  is related to scattering coefficient  $K$  by

$$\tau = NSK \quad (5)$$

In the case of nonabsorbing isotropic sphere,  $K$  can be expressed in a general form derived from Mie theory<sup>8,9)</sup> *i.e.*,

$$K = \frac{R}{\pi r^2} = \frac{2}{\alpha^2} \sum_{n=1}^{\infty} \frac{|a_n|^2 + |b_n|^2}{2n+1} \quad (6)$$

$$a_n = (-1)^n i(2n+1) \frac{S_n'(\beta) S_n(\alpha) - m S_n'(\alpha) S_n(\beta)}{S_n'(\beta) \phi_n(\alpha) - m \phi_n'(\alpha) S_n(\beta)},$$

$$b_n = (-1)^{n+1} i (2n+1) \frac{m S_n(a) S_n'(\beta) - S_n(\beta) S_n'(a)}{m \phi_n(a) S_n'(\beta) - S_n(\beta) \phi_n'(a)}$$

$$S_n(a) = \left( \frac{\pi a}{2} \right)^{1/2} J_{n+1/2}(a),$$

$$\phi_n(a) = \left( \frac{\pi a}{2} \right)^{1/2} [J_{n+1/2}(a) + (-1)^n i J_{-n-1/2}(a)],$$

$$n = 1, 2, 3, \dots,$$

$$\alpha = 2\pi r / \lambda^* \quad \beta = m\alpha,$$

$$= 2\pi r m_0(\lambda)^{-1}$$

The primes attached to  $S_n$  and  $\phi_n$  represent the derivatives and  $J$  represents the Bessel function of half order.  $r$  is the radius of sphere,  $\lambda^*$  the wavelength of the light in the surrounding medium,  $\lambda$  the wavelength in vacuum,  $m$  the relative refractive index and  $m_0$  the refractive index of medium.

For  $r \ll \lambda$ , eq. (6) reduces to well-known Rayleigh's equation,

$$K_R = \left( \frac{1}{\pi r^2} \right) 24 \pi^3 \left| \frac{m^2 - 1}{m^2 + 2} \right|^2 \frac{V^2}{\lambda^4} \quad (7)$$

where  $V$  is the volume of the sphere equal to  $4\pi r^3/3$ . Numerical computation of  $K$  in eq. (6) has been carried out for a variety of  $a$  and  $m$  by La Mer *et al.*<sup>10)</sup> Gumprecht and Sliepcevic,<sup>11)</sup> and Pangonis, Hell and Jacobson.<sup>12)</sup>

Figure 1 shows a typical example of the relation between  $K$  and  $a$  for the three values of  $m$  from their tables.

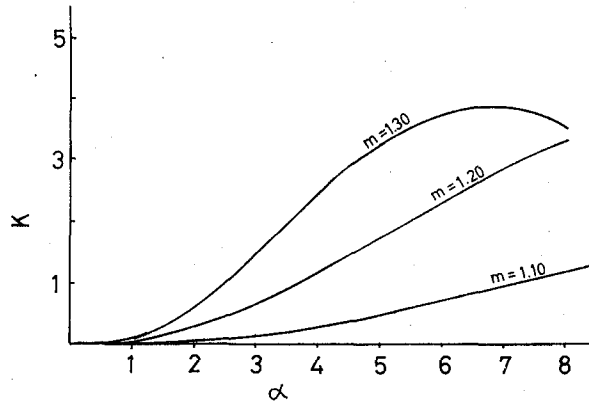


Fig. 1.  $K$  vs.  $\alpha$

Now, as is clear from eq. (6).  $K$  is a function of  $r$ ,  $m$ ,  $m_0$  and  $\lambda$ ; *i.e.*,

$$K \equiv f(a, \beta) \equiv F(a, m) \equiv G(m_0, m, r, \lambda) \quad (8)$$

So, it is proved that the turbidity ( $\tau$ ) is a function of  $N$ ,  $m_0$ ,  $m$ ,  $r$  and  $\lambda$ .

#### (b) Dispersion Quotient (DQ)

Many years ago, Teorell<sup>4)</sup> made an attempt to determine the particle size of colloidal dispersions by transmission measurements. In his article, it is suggested that the ratio

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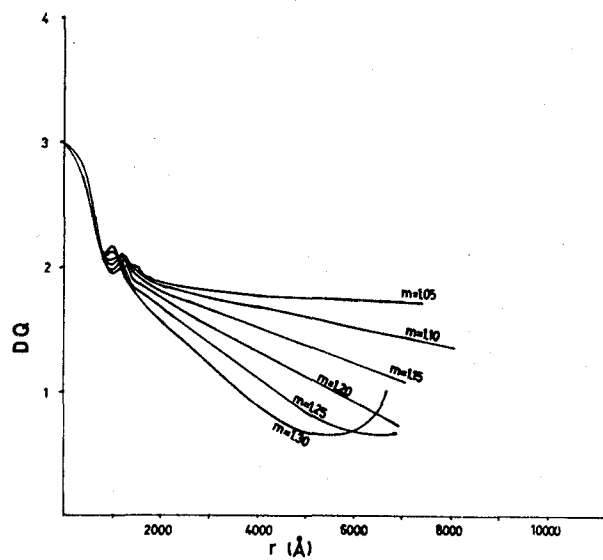


Fig. 2. Relation between dispersion quotient DQ and particle radius  $r$  for wavelengths 380 and 500  $m\mu$ .

$m$ : relative refractive index

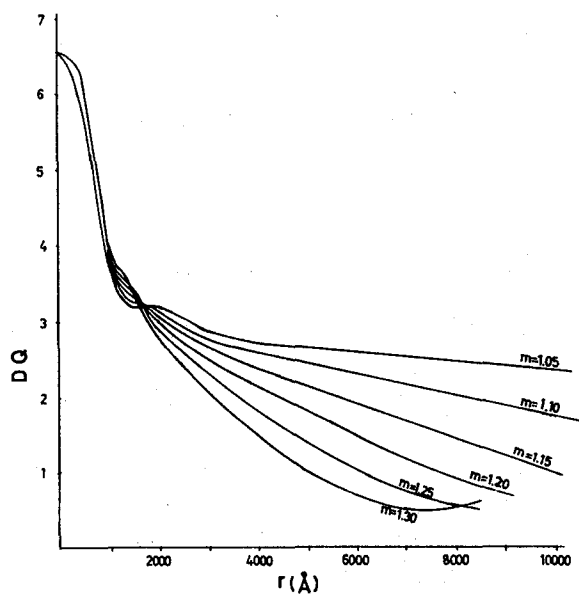


Fig. 3. Relation between dispersion quotient DQ and particle radius  $r$  for wavelengths 500 and 800  $m\mu$ .

$m$ : relative refractive index

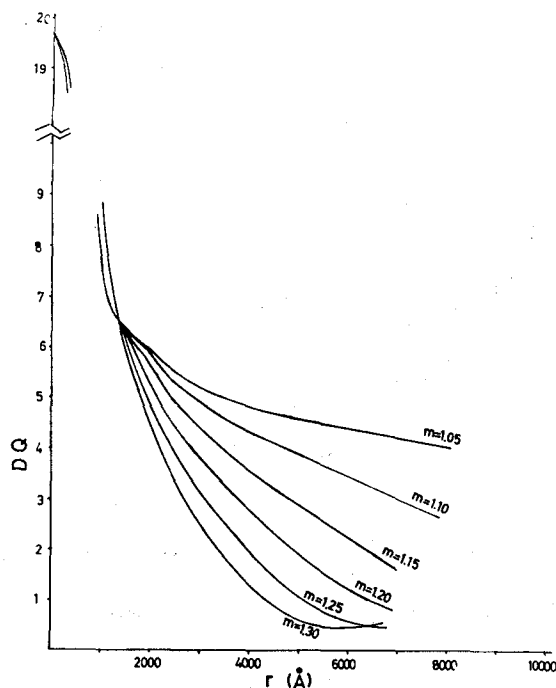


Fig. 4. Relation between dispersion quotient DQ and particle radius  $r$  for wavelengths 380 and 800  $m\mu$ .

$m$ : relative refractive index

of turbidities at two different wavelengths is to be related to particle size. The ratio is called "dispersion quotient" (DQ).

Now, as mentioned above, the ratio of turbidities, *i.e.*  $DQ(\lambda_1/\lambda_2)$ , measured at two different wavelengths ( $\lambda_1, \lambda_2$ ) can be represented in the following forms from eqs. (5) and (8).

$$\begin{aligned} DQ(\lambda_1/\lambda_2) &= \tau(\lambda_1)/\tau(\lambda_2) = K(\lambda_1)/K(\lambda_2) \\ &= G(m_0, m, r, \lambda_1) / G(m_0, m, r, \lambda_2) \end{aligned} \quad (9)$$

When the radius of sphere is very small in comparison with the wavelength (Rayleigh scattering), the equation (9) reduces to

$$DQ(\lambda_1/\lambda_2) = (\lambda_2/\lambda_1)^4 \quad (10)$$

with the aid of eq. (7). Hence, the quantity  $DQ(\lambda_1/\lambda_2)$  becomes independent of the particle radius. However, as shown in Figs. 2, 3 and 4, when the radius of sphere is in a same order of magnitude as the wavelength,  $DQ(\lambda_1/\lambda_2)$  becomes to be a function of  $r$  and  $m$ , for given two wavelengths  $\lambda_1$  and  $\lambda_2$ . Figures 2, 3 and 4 show the relations between DQ and  $r$ , which are obtained from tables I, II and III. Data in these tables were obtained from ratios of the  $K$  values computed by using Newton's interpolation formula for the table presented by Pangonis, Heller and Jacobson.<sup>12)</sup>

Also, these tables can be used to determine the radius of the particle by DQ-method.

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Table I. Dispersion Quotient (DQ) at 380 m $\mu$  and 500 m $\mu$ .

r(A)	m					
	1.05	1.10	1.15	1.20	1.25	1.30
400	2.730	2.756	2.782	2.804	2.824	2.840
500	2.567	2.592	2.613	2.634	2.654	2.671
600	2.388	2.402	2.414	2.423	2.429	2.435
800	2.084	2.079	2.076	2.077	2.085	2.106
1000	1.967	1.992	2.030	2.079	2.137	2.187
1200	1.999	2.046	2.077	2.074	2.030	1.943
1400	2.000	1.994	1.953	1.891	1.828	1.778
1600	1.932	1.899	1.861	1.824	1.782	1.727
1800	1.881	1.863	1.835	1.791	1.728	1.646
2000	1.865	1.846	1.803	1.735	1.645	1.537
2200	1.854	1.820	1.760	1.681	1.594	1.505
2400	1.837	1.794	1.730	1.647	1.535	1.412
2600	1.820	1.775	1.700	1.595	1.475	1.340
2800	1.809	1.755	1.671	1.561	1.424	1.253
3000	1.798	1.742	1.647	1.513	1.356	1.191
3200	1.794	1.726	1.612	1.471	1.299	1.099
3400	1.789	1.709	1.583	1.430	1.229	1.015
3600	1.774	1.690	1.560	1.387	1.179	0.955
3800	1.773	1.678	1.525	1.336	1.107	0.890
4000	1.764	1.660	1.496	1.297	1.056	0.820
4200	1.758	1.642	1.466	1.245	0.990	0.780
4400	1.745	1.624	1.437	1.201	0.939	0.739
4600	1.739	1.608	1.405	1.154	0.882	0.696
4800	1.737	1.593	1.371	1.105	0.825	0.655
5000	1.726	1.574	1.343	1.059	0.783	0.625
5200	1.722	1.557	1.308	1.015	0.739	0.603
5400	1.717	1.541	1.277	0.964	0.696	0.581
5600	1.710	1.521	1.242	0.924	0.669	0.592
5800	1.704	1.504	1.210	0.875	0.640	0.604
6000	1.700	1.484	1.171	0.835	0.621	0.654
6200	1.692	1.466	1.141	0.794	0.607	0.720
6400	1.687	1.446	1.100	0.754	0.602	0.823
6600	1.682	1.426	1.054	0.718	0.605	0.962
6800		1.408		0.688		
7000		1.386				
7250		1.361				
7500		1.335				
7750		1.307				
8000		1.282				
8500		1.227				
9000		1.172				
9500						

Table II. Dispersion Quotient (DQ) at 500  $m\mu$  and 800  $m\mu$ .

r(A)	m					
	1.05	1.10	1.15	1.20	1.25	1.30
400	6.073	6.137	6.196	6.266	6.335	6.399
500	5.753	5.834	5.910	5.991	6.067	6.134
600	5.386	5.474	5.558	5.637	5.710	5.777
800	4.618	4.677	4.728	4.775	4.815	4.853
1000	3.915	3.926	3.935	3.952	3.961	3.988
1200	3.436	3.445	3.466	3.510	3.576	3.675
1400	3.224	3.280	3.358	3.452	3.531	3.580
1600	3.191	3.278	3.351	3.381	3.371	3.301
1800	3.198	3.257	3.269	3.244	3.171	3.088
2000	3.168	3.165	3.098	2.998	2.879	2.799
2200	3.106	3.076	3.008	2.837	2.712	2.517
2400	3.033	2.977	2.876	2.733	2.579	2.426
2600	2.962	2.888	2.778	2.646	2.498	2.324
2800	2.900	2.824	2.716	2.568	2.401	2.197
3000	2.852	2.777	2.666	2.498	2.302	2.057
3200	2.821	2.747	2.617	2.427	2.188	1.950
3400	2.802	2.716	2.557	2.334	2.100	1.841
3600	2.784	2.677	2.494	2.249	2.013	1.728
3800	2.761	2.635	2.446	2.186	1.909	1.596
4000	2.736	2.602	2.400	2.120	1.825	1.511
4200	2.716	2.574	2.349	2.062	1.743	1.378
4400	2.705	2.548	2.300	2.000	1.651	1.275
4600	2.688	2.520	2.255	1.932	1.566	1.182
4800	2.669	2.490	2.111	1.869	1.481	1.099
5000	2.663	2.465	2.160	1.794	1.390	1.014
5200	2.649	2.435	2.111	1.729	1.317	0.945
5400	2.631	2.405	2.066	1.671	1.245	0.866
5600	2.621	2.377	2.018	1.602	1.159	0.797
5800	2.607	2.351	1.975	1.538	1.080	0.745
6000	2.592	2.326	1.929	1.472	1.009	0.689
6200	2.585	2.299	1.876	1.405	0.952	0.641
6400	2.574	2.271	1.830	1.350	0.886	0.591
6600	2.562	2.243	1.787	1.286	0.825	0.556
6800	2.549	2.213	1.738	1.222	0.773	0.532
7000	2.533	2.183	1.694	1.166	0.727	0.506
7250	2.518	2.149	1.636	1.091	0.673	0.484
7500	2.510	2.115	1.574	1.028	0.623	0.476
7750	2.504	2.083	1.520	0.953	0.578	0.471
8000	2.482	2.045	1.460	0.897	0.550	0.502
8500	2.459	1.974	1.337	0.779	0.501	0.595
9000		1.901				
9500		1.828				
10000		1.752				
11000		1.601				
12000		1.452				



Estimation of Particle Size, Refractive Index and Concentration of Latices

Table III. Dispersion Quotient (DQ) at 380 m $\mu$  and 800 m $\mu$ .

r(A)	m					
	1.05	1.10	1.15	1.20	1.25	1.30
400	16.58	16.91	17.24	17.57	17.89	18.17
500	14.77	15.12	15.45	15.78	16.10	16.39
600	12.86	13.15	13.41	13.66	13.87	14.07
800	9.622	9.723	9.815	9.917	10.04	10.22
1000	7.699	7.819	7.989	8.217	8.463	8.723
1200	6.868	7.049	7.198	7.279	7.261	7.140
1400	6.447	6.539	6.557	6.528	6.456	6.366
1600	6.164	6.224	6.237	6.167	6.008	5.701
1800	6.014	6.066	5.999	5.808	5.479	5.081
2000	5.905	5.843	5.586	5.202	4.734	4.302
2200	5.759	5.599	5.295	4.769	4.322	3.789
2400	5.571	5.340	4.975	4.502	3.958	3.425
2600	5.391	5.127	4.724	4.221	3.684	3.113
2800	5.246	4.957	4.539	4.009	3.420	2.752
3000	5.129	4.838	4.391	3.781	3.120	2.449
3200	5.061	4.743	4.220	3.569	2.843	2.144
3400	5.012	4.642	4.047	3.338	2.583	1.872
3600	4.938	4.525	3.889	3.118	2.373	1.649
3800	4.893	4.422	3.730	2.920	2.131	1.420
4000	4.824	4.319	3.591	2.749	1.926	1.239
4200	4.773	4.226	3.442	2.568	1.725	1.074
4400	4.720	4.138	3.305	2.403	1.550	0.941
4600	4.675	4.052	3.168	2.230	1.381	0.823
4800	4.636	3.966	3.033	2.066	1.222	0.720
5000	4.595	3.880	2.901	1.900	1.088	0.633
5200	4.561	3.791	2.760	1.754	0.973	0.569
5400	4.517	3.705	2.638	1.611	0.867	0.503
5600	4.482	3.616	2.507	1.481	0.774	0.471
5800	4.441	3.535	2.390	1.346	0.692	0.450
6000	4.407	3.453	2.259	1.229	0.631	0.451
6200	4.373	3.371	2.141	1.115	0.578	0.462
6400	4.341	3.285	2.013	1.019	0.534	0.487
6600	4.308	3.199	1.884	0.923	0.499	0.535
6800		3.114		0.841		
7000		3.027				
7250		2.925				
7500		2.823				
7750		2.723				
8000		2.620				
8500		2.421				
9000		2.228				
9500						

(c) **Method for Determination of Particle Radius and Relative Refractive Index from Turbidity Measurements**

Suppose that turbidities,  $\tau(\lambda_1)$ ,  $\tau(\lambda_2)$ ,  $\tau(\lambda_3)$  and  $\tau(\lambda_4)$  are given for the same test solution by the transmission measurements at the four different wavelengths,  $\lambda_1$ ,  $\lambda_2$ ,  $\lambda_3$  and  $\lambda_4$ , and that the refractive index of medium ( $m_0$ ) is known, then, the following relations will be obtained from eq. (9), for the same refractive index of particle relative to surrounding medium and the same radius( $r$ ).

$$\begin{aligned}\tau(\lambda_1)/\tau(\lambda_2) &= DQ(\lambda_1/\lambda_2) = G(m_0, m, r, \lambda_1) / G(m_0, m, r, \lambda_2) \\ &\equiv G_1^*(m, r)\end{aligned}\quad (11)$$

$$\begin{aligned}\tau(\lambda_3)/\tau(\lambda_4) &= DQ(\lambda_3/\lambda_4) = G(m_0, m, r, \lambda_3) / G(m_0, m, r, \lambda_4) \\ &\equiv G_2^*(m, r)\end{aligned}\quad (12)$$

In above equations,  $\tau(\lambda_1)/\tau(\lambda_2)$  and  $\tau(\lambda_3)/\tau(\lambda_4)$  are known and,  $G_1^*$  and  $G_2^*$  are certain function of  $m$  and  $r$ . Therefore, one should be able to estimate a solution for  $m$  and  $r$  from eqs. (11) and (12). Though these equations can not be solved algebraically because of the complication of  $G^*$ , they can be done graphically. The way of the graphical solution is as follows. Firstly, estimate the relation between  $m$  and  $r$  for a given  $DQ(\lambda_1/\lambda_2)$  with aid of a plot of  $DQ$  against  $r$  with various relative refractive index ( $m$ ), such as Figs. 2-4. Next, carry out similar operation for another given  $DQ(\lambda_3/\lambda_4)$ . Then, two curves(or relations) obtained here are expected to intersect at a point, for example at  $m=m^*$  and  $r=r^*$ , if eqs. (12) and (13) have a real solution.  $m^*$  and  $r^*$  are a required solution for these equations.

Two distinct dispersion quotients can be sufficiently determined from turbidities measured at least at three different wavelengths. In this paper, these quotients were measured at three different wavelengths,  $\lambda_1=380\text{ m}\mu$ ,  $\lambda_2=500\text{ m}\mu$  and  $\lambda_3=800\text{ m}\mu$ .

(d) **Method for Determination of Concentration from Turbidity Measurements**

The number of particles per unit volume( $N$ ) can be transformed to the concentration (c) (g/100 cc) by the following equation.

$$NVd = N\left(\frac{4}{3}\pi r^3\right)d = c \times 10^2 \quad (13)$$

Here,  $V$  is the volume of a particle and  $d$  is the density. Substitution of eq. (13) into eq. (5) results

$$c = 3.0701 \times 10^2 \times \left(\frac{r \cdot d}{K \cdot l}\right) \times \log(I_0/I) \quad (14)$$

where  $l$  is the optical path length in the cell of an apparatus used for the measurements and  $(I/I_0)$  the light transmission. In the right hand of eq. (14), the radius of particle( $r$ ) and the relative refractive index( $m$ ) are known by the preceding method. Thus,  $K$  values corresponding to  $m$  can be calculated by using the relation between  $K$  and  $\alpha$  in the table, if the refractive index of medium( $m_0$ ) and the wavelength are given. Through these procedures, the concentration of particle can be estimated by eq. (14), when the density( $d$ ) is known.

## EXPERIMENTAL

## (a) Materials

Measurements were carried out on a monodispersed polystyrene latex obtained from Dow Chemical Company. This sample was labeled that the diameter was  $0.481 \mu$  with the standard deviation of  $1.8 \times 10^{-3} \mu$ . The concentration of original solution used in this study was 0.2172 g per 100 ml. Test solution was prepared by diluting 2 ml of the original solution with 100 ml pure water.

Since other adequate latices of polystyrene were not available only one sample mentioned above was used in this study.

## (b) Measurement of Light Transmission

Transmission measurements were carried out at  $20^\circ\text{C}$  with photometer, Type SP-88 from Simazu Seisakusho Co. The measurements were made with light beam of three different wavelengths;  $380 \text{ m}\mu$ ,  $500 \text{ m}\mu$  and  $800 \text{ m}\mu$ , using six matched crystal cells. The depths of these cells were matched with 1.000 cm.

## (c) Wavelength Dependence on Refractive Index

Refractive index generally depends on wavelength. In the present work, as the refractive indices of pure water the values of 1.3460, 1.3364 and 1.3286 were adopted for wavelengths  $380 \text{ m}\mu$ ,  $500 \text{ m}\mu$  and  $800 \text{ m}\mu$ , respectively. Here, it was assumed that the relative refractive index of polystyrene was independent of wavelengths in the range from  $380 \text{ m}\mu$  to  $800 \text{ m}\mu$ . But actually it was varied from 1.21 to 1.19 in that range of wavelength.<sup>13,14)</sup>

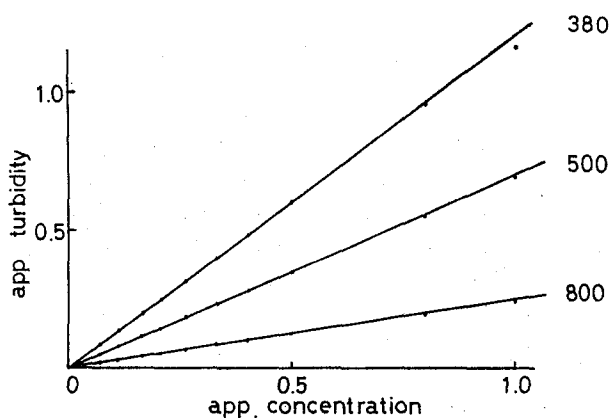
## RESULTS

## (a) Results of Turbidity Measurements

Table IV and Fig. 5 show the results of turbidity measurements for the test solution. The concentration of the solution was defined to be unity for convenience and denoted

Table IV. Apparent Turbidity for Various Apparent Concentrations.

apparent conc	$380 \text{ m}\mu$		$500 \text{ m}\mu$		$800 \text{ m}\mu$	
	I	$\tau'$	I	$\tau'$	I	$\tau'$
1.000	6.8	1.168	20.0	0.699	57.0	0.244
0.803	11.2	0.951	27.2	0.565	63.6	0.197
0.500	24.7	0.607	44.4	0.352	75.0	0.125
0.402	32.5	0.488	51.7	0.287	79.6	0.0991
0.333	39.1	0.408	58.1	0.236	82.7	0.0825
0.268	46.6	0.332	64.2	0.193	58.8	0.0665
0.200	57.0	0.244	72.1	0.142	89.3	0.0491
0.161	63.5	0.197	77.1	0.113	91.2	0.0400
0.125	71.0	0.149	81.3	0.0899	93.0	0.0315
0.080	79.9	0.0975	88.0	0.0555	95.5	0.0200

Fig. 5. Apparent turbidity *vs* apparent concentration.

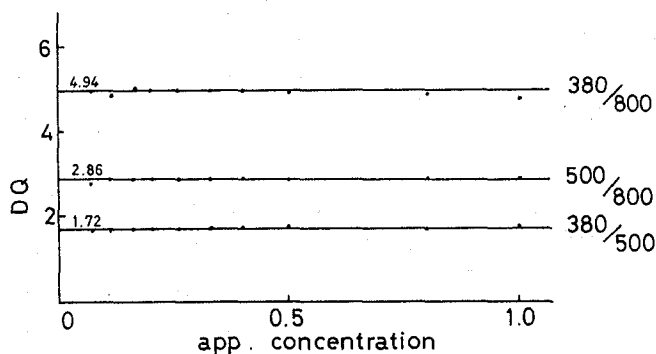
as "apparent concentration" ( $c'$ ). The apparent turbidity  $\tau'$  is  $\tau L$ . It is seen in Fig. 5 that the relation between  $\tau'$  and  $c'$  is linear in the range of  $c'$  smaller than 0.8, where the Lambert-Beer equation is valid.

#### (b) Determination of DQ Value

Table V and Fig. 6 show DQ values calculated from Table IV for various apparent concentrations.

Table V. DQ value for Various Concentrations, Obtained from Table IV.

apparent conc.	DQ 380/500	DQ 500/800	DQ 380/800
1.000	1.670	2.864	4.783
0.803	1.682	2.877	4.838
0.500	1.722	2.823	4.862
0.402	1.704	2.891	4.925
0.333	1.729	2.858	4.943
0.268	1.723	2.895	4.986
0.200	1.722	2.891	4.967
0.161	1.745	2.823	4.929
0.125	1.654	2.852	4.718
0.080	1.756	2.750	4.873

Fig. 6. DQ *vs* apparent concentration.

From Table V and Fig. 6, it is seen that DQ varies slightly with change in the concentration. This concentration dependence of DQ value seems to come from the secondary light scattering of particles. Accordingly, the intrinsic DQ value ( $DQ_{c \rightarrow 0}$ ) was determined by the following procedures. Method 1; as mentioned above, the relation between  $\tau'$  and  $c'$  forms a straight line passing through origin for the dilute solution. Then,  $DQ_{c \rightarrow 0}$  can be determined from the ratio of the gradients of the two straight lines. Method 2; one can estimate  $DQ_{c \rightarrow 0}$  by extrapolating  $c'$  to zero in the DQ- $c'$  relation.

The intrinsic DQ values obtained by Method 1 were  $DQ(380/500)_{c \rightarrow 0} = 1.70$ ,  $DQ(500/800)_{c \rightarrow 0} = 2.84$  and  $DQ(380/800)_{c \rightarrow 0} = 4.84$  respectively, and those obtained by Method 2 were  $DQ(380/500)_{c \rightarrow 0} = 1.72$ ,  $DQ(500/800)_{c \rightarrow 0} = 2.86$  and  $DQ(380/800)_{c \rightarrow 0} = 4.94$ .

### (c) Determination $r$ and $m$

The relation between  $r$  and  $m$  at a given  $DQ_{c \rightarrow 0}$  can be obtained by using the DQ- $r$  relation with various  $m$ , such as Figs. 2-4. Table VI and Fig. 7 show the  $r$ - $m$  relations for three  $DQ_{c \rightarrow 0}$  values determined by Method 1. From Fig. 7, it is found that three  $m$ - $r$  curves intersect each other at a same point. The co-ordinates of this point give the required values of  $r$  and  $m$ .  $r = 2280 \text{ \AA}$  and  $m = 1.19$  were obtained from the  $DQ_{c \rightarrow 0}$  values determined by Method 1. For those determined by Method 2,  $r$  and  $m$  were  $2200 \text{ \AA}$  and  $1.18$ , respectively.

Table VI. Radius of Particle obtained from DQ for various  $m$ .

$m$	DQ 380/500 1.70	DQ 500/800 2.84	DQ 380/800 4.84
1.05	6000	3100	3900
1.10	3500	2760	3020
1.15	2600	2480	2500
1.20	2140	2210	2180
1.25	1860	2020	1950
1.30	1670	1890	1810

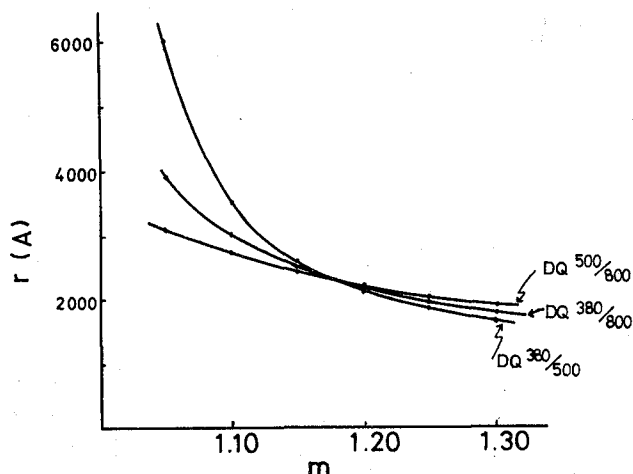


Fig. 7. Relation between particle radius  $r$  and relative refractive index  $m$ , obtained from DQ.

On the other hand, the radius of the latex determined by the observation with a electron microscopy was 2350–2450 Å. The relative refractive indices settled by Bateman, Weneck and Eshler,<sup>14)</sup> were 1.19–1.21 within the wavelengths used in this work.

#### (d) Determination of the Concentration

The calculation of  $c$  by eq. (15) was made for  $\lambda=500$  m using  $m_o=1.336$ ,  $d=1.06$ ,  $I=1.336$  and  $\log(I_o/I)=0.710$ .  $C$  was 0.2122 in g per 100 ml, for  $m$  and  $r$  determined by Method 1. For those determined by Method 2,  $c$  was 0.2312 in g per 100 ml. On the other hand,  $c$  determined by drying 50 ml of original solution at 40°C in vacuum, was 0.2172 in g per 100 ml.

### DISCUSSION

Results are summarized in Table VII. From this Table, it is seen that  $r$ ,  $m$  and  $c$  determined by the present method are in good agreement with those obtained by other methods. However, it seems that the effectiveness of the present method for the general use is somewhat restricted by some factors discussed below.

Table VII. Summary of latex radius, relative refractive index and concentration values.

method	$r(\text{\AA})$	$m$	$c$ (g/100 ml)
our method	1 2280	1.19	0.2122
	2 2200	1.18	0.2312
electron microscopy	2350 ~ 2450	—	—
dry weight	—	—	0.2172
reference	2405 <sup>a)</sup>	1.19 ~ 1.21 <sup>b)</sup>	0.2 <sup>a)</sup>

a) labeled by Dow Chemical Company

b) data of Bateman, Weneck and Eshler<sup>13)</sup>

#### (a) Measurable Range of $r$

From Figs. 2–4, it is observed that a DQ- $r$  relation gives two or more radii for a value of DQ and  $m$ , when  $r$  is in an order of 1500 Å and less, or more than 8000 Å. These mean that, in these ranges of  $r$ , one can not determine unequivocally the  $m$ - $r$  relation from DQ- $r$  relations with  $m$  and a DQ value. Accordingly, it will be reasonable to judge that the measurable range of  $r$  by the present method is roughly from 1500 Å to 8000 Å, although this range may be changed by the choice of wavelength for the turbidity measurement.

#### (b) Wavelength Dependence on $m$

The theory of present method is established by assuming  $m$  to be constant for any wavelengths. Fortunately, the wavelength dependence on  $m$  was very minor in practice, when the range of wavelength used to measure the turbidities is not so large. Consequently, the present method can be successfully applied to determinate  $r$ ,  $c$  and  $m$  at a same time, within a practically allowable limit of error.

When  $m$  depends strongly upon wavelength, it is necessary to take account of wavelength dependence into the tabulation of DQ values related to  $m$ . Such further theoretical treatments will be achieved in another opportunity.

### (c) Polydispersity of Particles

In this paper, the method of measurement is given for monodisperse particles. Accordingly, it is desirable to check the validity of this method for systems containing polydisperse particles. In other words, it is necessary to confirm theoretically and experimentally how average values of  $r$ ,  $m$  and  $c$  are obtained by this method, when the system is polydisperse. Concerning with this problem, it is prearranged to carry out experimental check with mixtures of two or more distinct monodisperse polystyrene latices in near future.

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